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STRUCTURAL EFFECTS IN THE DIELECTRIC CONSTANT OF RARE-EARTH OXIDES: Nd₂O₃

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ABSTRACT

Thin films of Nd_2O_3 have been studied. For the amorphous phase the dielectric constant is ~ 11 whilst the refractive index is 1.76 ± 0.02 , in the cubic form these values are $\sim 13.6 - 15.4$ and 1.93 ± 0.02 . Hexagonal films have dielectric constants in the range 17 - 21 and estimates based upon the cubic values lead to $k \sim 27$, we were unable to determine a reliable refractive index value. The normally high temperature hexagonal phase could be nucleated on a Si substrate by deposition at relatively low temperature ~ 280 °C.

INTRODUCTION

In standard metal-oxide-semiconductor field effect transistors (MOSFETs), when the gate oxide thickness is reduced to a few atomic layers, the gate leakage surpasses the transistor-off current leakage (1). It is consequently recognized that for device scaling to continue, a change in the transistor composition to include higher dielectric insulators than SiO_2 will be necessary (2) so that thicker dielectric films can be used with, presumably, lower leakage currents. The hypothesis here, of course, is that a dielectric of thickness $d_{dielectric}$ having a dielectric constant, k, can straightforwardly replace an SiO_2 film of thickness d_{SiO_2} providing $d_{dielectric}/d_{SiO_2} = k/3.9$. For this application various binary and ternary oxides have been studied over the past few years and the rare earth (RE) oxides are amongst the materials examined. The primary reason for the examination of RE_2O_3 oxides is that they appear to have k values > 3.5 times the value for SiO_2 . There are, however, certain associated problems with the RE_2O_3 oxide series such as a) potential interaction of the film with the silicon surface (3) which leads to silicide formation or a thin SiO_2 layer due to segregation of the oxide into the metal and oxygen, b) the films are hygroscopic c) anisotropy of the dielectric constant may be relevant for

polycrystalline oxides d) requirement of a new metallic gate electrode and e) strongly ion dependent dielectric constant. Despite the problems research into these oxides remains popular because they can have interestingly high dielectric constants, low leakage currents and a reasonable optical band gap (4).

As a general rule the RE oxides based on elements with atomic number Z > 62 form in a cubic crystalline phase with a dielectric constant which may range from 12.5 to 14.3 (5). It has been demonstrated that for those oxides based on the RE elements with an atomic number Z < 62, the so called "light" rare-earths, two crystalline phases can be obtained (cubic and hexagonal) and in consequence two different values of dielectric constant are anticipated. A good example for a RE oxide with Z < 62 is Pr_2O_3 while for Z > 62 it is Pr_2O_3 in its cubic phase also has a dielectric constant of 15 (6), which increases to 14 (5). Pr_2O_3 in its cubic phase also has a dielectric constant of 15 (6), which increases to 23-25 (7) in the hexagonal phase (as observed for polycrystalline samples (5,7)). The hexagonal phase is stabilized only for films annealed at 750 °C when deposited onto Pr_2O_3 has a higher dielectric constant than Pr_2O_3 is not, in fact, surprising since it has been demonstrated (5) that the molecular volume for the hexagonal phase is smaller than for the cubic phase leading directly to a larger molar permittivity.

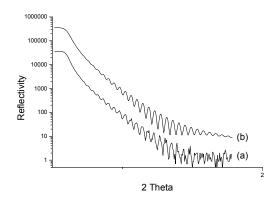
In the present work we have chosen to try to complete our understanding of the electrical and optical properties of the RE_2O_3 oxides by examining the case of Nd_2O_3 . Nd_2O_3 is likely to be very similar to Pr_2O_3 , because it has both the cubic and hexagonal phases and is close in atomic number. In the cubic form Nd_2O_3 is reported to have a dielectric constant in the range 12.9 to 13.9 (6,8) close to the value of 15 measured for the $c\text{-}Pr_2O_3$. The dielectric constant of the hexagonal phase of Nd_2O_3 has not been reported and it is partly the purpose of the present work to try to "fill in" this information gap and confirm our hypothesis about the importance of molecular volume for the RE_2O_3 compound series.

EXPERIMENT

Thin films of Nd₂O₃ were deposited onto p - type Si(100) 10-15 ohm cm resistivity wafers following an HF rinse, via electron beam evaporation. The starting material was Nd₂O₃ single crystal. A partial pressure of 2.5 x 10⁻⁵ torr of oxygen was introduced in the chamber to ensure the stoichiometry of the deposited film. Three different sequences of sample processing were used: the first deposition, at room temperature, at a deposition rate of 2 Å s⁻¹, the second involved heating the sample holder to a temperature of 280 °C and using same deposition rate of 2 Å s⁻¹ and the third, still at 280 °C, but with a lower deposition rate of 0.4 Å s⁻¹. A total thickness, measured in-situ using a quartz oscillator, and confirmed by ex-situ ellipsometry at a wavelength of 632.8 nm, of 88 nm was typically used. The three different types of samples were also annealed at 600 °C for 3 hours in O₂ atmosphere and then at 800 °C for 30 minutes in an N₂ atmosphere. The N₂ gas, for high temperature annealing, was used to minimize possible oxidation of the Si at the dielectric /Si interface while the lower temperature O₂ anneal was used to ensure reoxidation the Nd₂O₃ if by chance it was sub-stoichiometric in the as-deposited state.

Oxide film densities and roughness were determined by glancing angle x-ray reflectivity using a Philips double crystal diffractometer with Cu- $K_{\alpha 1}$ radiation (λ =

0.1541 nm) together with a four crystal Bartels monocromator and 0.45 mm receiving slit. Typical experimental data are shown in Figs.1,2 and 3 including the fits obtained using the WINGIXA software supplied by PANanalytical company. Note that the fit curves have been displaced vertically so that the reader can more clearly visualize the closeness of the fit to the experimental data. For the amorphous samples (as-deposited at room temperature), an average density value of 6.4 ± 0.2 g cm⁻³ was obtained. For the samples that were annealed at 600 °C the density was 6.70 ± 0.2 g cm⁻³ and a density value of 7.02 ± 0.2 g cm⁻³ was obtained for the sample that received the further annealing treatment at 800 °C. From the JADE database (9), the cubic and the hexagonal densities are expected to be respectively 6.57 and 7.16 g cm⁻³.



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Fig. 1 Glancing incidence X ray reflectivity data for amorphous Nd₂O₃ deposited at room temperature at 2 Å/sec. Curve (a) represents the raw data and curve (b) is the fit performed with the WINGIXA software. The simulation indicates that the samples have a density of 6.4 ± 0.2 g cm⁻³ and a roughness of 1.5 nm.

Fig.2 Glancing incidence X ray reflectivity data for Nd_2O_3 deposited at 280 °C at 2 Å /sec. followed by 600 °C annealing for 3h in an O_2 atmosphere. Curve (a) represents the raw data and the (b) curve is the fit performed with the WINGIXA software. The simulation indicates that the samples have a density of 6.7 ± 0.2 g cm⁻³ and a roughness of 1.1 nm.

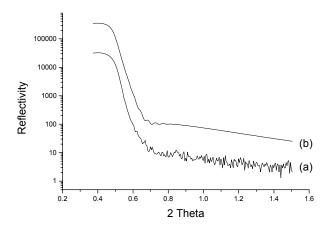


Fig.3 Glancing incidence X ray reflectivity data for Nd_2O_3 deposited at 280 °C at 0.4 Å/sec. followed by 600 °C annealing for 3h in O_2 atmosphere and then 800 °C for 30 min in N_2 . Curve (a) represent the raw data and curve (b) is the fit performed with the WINGIXA software. The simulation indicates that the samples have a density of 7.02 ± 0.2 g cm⁻³ and a roughness of 3.9 nm.

Film textures were analyzed by X-ray diffraction (XRD) measured in the theta-2 theta mode using a Cu target with a current of 35 mA an acceleration voltage of 40 KV. In Figs.4, 5 and 6 we show the experimental diffraction patterns. On each line we have indicated the corresponding crystal plane. When a letter "c" or an "h" is present before that number, it indicates that that line is associated respectively with a cubic phase or hexagonal phase in the film. We underline the fact that for some samples both cubic and hexagonal phases are present in the oxide simultaneously.

The dielectric constant and the leakage current characteristics were obtained by performing capacitance/voltage C(V) and current/voltage I(V) measurements using a Keithley 590 system, the capacitance was measured at a frequency of 100 kHz. Metal oxide semiconductor (MOS) capacitors were manufactured by depositing 0.00196 cm² Al dots on the oxide film surface. For the sample deposited at room temperature, which appears amorphous from the X-Ray diffraction patterns we measured a dielectric constant(current density) in the range 11-12 (10⁻⁷ A cm⁻² at 1 MV cm⁻¹) whereas for the samples that were mainly composed of c-Nd₂O₃ we obtained 13-15 (10⁻⁴ A cm⁻² at 1 MV cm⁻¹). The samples which were a mixture of c-Nd₂O₃ and h-Nd₂O₃ indicated a dielectric constant in the range 17-18 and a current density of 10⁻⁴ A cm⁻².

Fourier transform infrared absorption (FTIR) spectra, in the absorbance mode using normal incidence or oblique incidence, were measured in the range of 400-1500 cm $^{-1}$. The primary peak for Nd₂O₃ was lower then 400 cm $^{-1}$ and therefore not accessible for our instrument. However, as demonstrated previously (10), FTIR can be used to semi-quantitatively estimate the amount of interfacial SiO₂ (if present) by observation of the transverse optic asymmetric stretch mode appearing at ~ 1070 cm $^{-1}$ (depending upon film thickness) or the longitudinal optic mode at ~ 1260 cm $^{-1}$. This measurement was carried out for the three types of samples and no evidence was found for any significant SiO₂ layer at the dielectric/Si interface. We quantify this to mean that any such interlayer was less than 1 nm thick.

DISCUSSION

Room Temperature Deposited Nd₂O₃

The sample deposited at room temperature was determined to be amorphous on the basis of the absence of a clear XRD line structure. It was subsequently subjected to a series of anneals in flowing O_2 in the temperature range $300-500\,^{\circ}\text{C}$ for 45 minute periods. No diffraction peaks were observed following these anneals. However, annealing at $600\,^{\circ}\text{C}$ did result in the appearance of diffraction peaks so the sample was then annealed at this temperature for 3 hours after which the structure of cubic polycrystalline Nd_2O_3 clearly appeared in the XRD pattern (Fig.4). We note, in passing, that the diffraction lines at 34.45° (411) and at 41.77° (431) are barely perceptible above the level of the noise. In the absence of these lines, the XRD pattern could have been misinterpreted as being that of cubic NdO_2 since it presents a similar pattern as c- Nd_2O_3 , but slightly shifted by 0.4° towards smaller 2 theta. Such a range of shift could be the result of the alignment of the instrument.

As mentioned previously, we measured the density of the sample deposited at room temperature and then annealed at 600° C (Fig.1). The density value from our fitting, 6.7 ± 0.2 g cm⁻³, is consistent with but slightly larger than the cubic phase value quoted in the JADE database (9) of 6.57 g cm⁻³, and lower then the value of 6.97 g cm⁻³ reported for bulk c-NdO₂. This indirect measurement gives further credence to the assumption that the 600 °C annealed sample was indeed polycrystalline, cubic Nd₂O₃.

The measured refractive index of the as-deposited material was 1.76 ± 0.02 and the dielectric constant 11-11.8. This latter value is slightly smaller than a previously reported amorphous value (12.9, reference 6) and this suggests that our films may be less dense than those obtained by others. Following annealing at 600 °C dielectric constant increased to 13.6-15.4 and the refractive index increased to 1.93 ± 0.02 . Finally, after annealing at 800 °C in N_2 , the refractive index stabilized at 1.92 ± 0.02 .

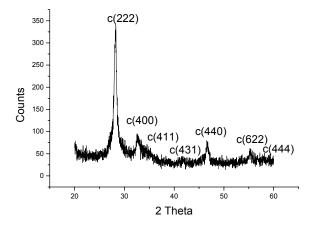


Fig. 4 X ray diffraction pattern for the Nd₂O₃ deposited at room temperature at 2 Å /sec. Then annealed at 600 °C for 3h in O₂. We identify the cubic phase related X ray lines as c(nnn), where (nnn) is the crystallographic plane. The silicon peaks are not present, because the sample was slightly tilted with respect to the incidence direction of the X-ray beam. The sample is polycrystalline.

Nd₂O₃ Deposited at 280 °C at 2 Å s⁻¹

The X ray diffraction spectra shown in Fig. 5 indicate that the as-deposited films contain both cubic and hexagonal phases of Nd₂O₃. It is not possible for us to estimate the relative quantities of each phase or if there is a significant amount of amorphous material. Following annealing at 600 °C in oxygen the density was ascertained to be 6.5 ± 0.2 g cm⁻³ which is slightly lower than the value of 6.7 ± 0.2 g cm⁻³ value determined for the annealed, amorphous material. Furthermore, the measured refractive index of the 600 °C annealed films was 1.82 ± 0.02 which is significantly lower than the value of 1.93 ± 0.02 observed for similarly annealed films which were initially deposited at room temperature. Following a further anneal at 800 °C the refractive index stabilized at 1.84 \pm 0.02, again smaller than the value for the annealed, initially amorphous films. The overall behavior of the density and the refractive index strongly points to the conclusion that deposition at 2 Å s⁻¹ combined with the temperature effect (280 °C substrate temperature) results in less dense films, perhaps columnar or porous, than we obtain if the initial films are deposited at room temperature. It is very interesting that deposition at 280 °C resulted in films which already contained some crystalline phase whereas heating the amorphous deposited films to substantially higher temperatures (500 °C for example) did not result in the appearance of crystallinity. Furthermore, it has been reported that nano-powders of Nd_2O_3 are cubic at 600 °C but transform to hexagonal in the 850 - 1000°C range (11) suggesting that high temperature treatment should lead to formation of the hexagonal phase. Annealing at 800 °C as shown in Fig. 5 appears to remove/eliminate the hexagonal component observed in 600 °C annealed films.

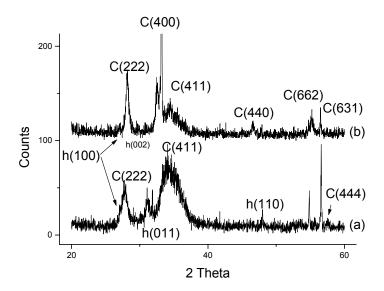


Fig. 5 X ray diffraction pattern for Nd₂O₃ deposited at 280 °C at 2 Å /sec. (a) followed by 600 °C annealing for 3h in O₂ and then at 800 °C for 30 min in N₂ (b). The X ray lines are identified with the different phases of the oxide: h is for hexagonal. c is for cubic. We also indicate the position of the Si(200) which comes from the substrate (upper curve). In the bottom curve the sample was tilted with respect to the incidence direction of the X-ray to demonstrate that the sample is polycrystalline and to minimize interference from the substrate diffraction pattern. The patterns indicate that the Nd₂O₃ is a single, cubic phase after the annealing.

Nd₂O₃ Deposited at 280 °C at 0.4 Å s⁻¹

The physical character of these films is different from the two previously studied materials. The as-deposited films show clear evidence for the hexagonal phase with a refractive index of 1.83 ± 0.02 and a dielectric constant of $12 - 12.7 \pm 0.2$. The refractive index of hexagonal Nd₂O₃ is expected to be ~ 2.11 (12) suggesting that the film is indeed composed of amorphous and hexagonal material. Assuming a heterogeneous mixture with an amorphous material of index 1.76 as we found above, we estimate the film would be composed of 80% amorphous phase and 20% hexagonal. Note, however, that the amorphous values were determined for room temperature deposited films whilst the films discussed here were deposited at 280 °C.

Upon annealing at 600 °C in oxygen followed by 800 °C in N_2 , the refractive index increases to 1.91-1.94 \pm 0.02 and the density is 7.02 \pm 0.2 g cm⁻³. Note that the refractive index is still smaller than the bulk value for the hexagonal phase (2.11) and the density is somewhat smaller than the bulk value (7.14 g cm⁻³) though within the error limits. There is also some evidence in the X ray spectrum (Fig. 6) for some cubic phase since annealing appears to result in a cubic (222) peak. We also note that the film has a dielectric constant of 17-18 \pm 0.2. It is clear that the deposition temperature and rate has a significant effect on the structure of the as-deposited films and on their behavior as a function of annealing temperature.

Estimation of the Hexagonal Dielectric Constant

The primary objective of the work reported here was to obtain some estimate of the dielectric constant associated with the hexagonal phase of Nd_2O_3 . In order to do this we first examine refractive index data for the different films. The refractive index is related to the molecular volume, V_m , and the optical polarizability, α_{opt} , through the Lorentz-Lorenz equation (13):

$$(n^2 - 1)/(n^2 + 2) = 4 \pi \alpha_{\text{opt}}/3 V_{\text{m}}$$
 (1)

For the hexagonal phase we have n = 2.11 and V_m = 78 ų leading to α_{opt} = 9.96 ų. To first order we will assume that the polarizability does not depend upon molar volume so that we can calculate n_{cubic} by simply substituting V_m = 85 ų in Eq. 1. We therefore expect the cubic phase refractive index to be 1.97. The value measured for 600 °C annealed amorphous films was 1.93 ± 0.02 which is satisfactorily close to the estimated value allowing for the fact that potential molar volume effects have been ignored. We are therefore lead to conclude that the measured values of n = 1.93 ± 0.02 and k = 13.6 – 15.4± 0.02 are reasonable approximations to the correct values for the cubic phase of Nd₂O₃. We reiterate that the measured density of the annealed samples was 6.7 ± 0.2 g cm⁻³ which is also very close to the bulk, cubic Nd₂O₃ value of 6.57 g cm⁻³.

The refractive index and dielectric constant obtained for the thin film cubic Nd_2O_3 are consequently in satisfactory agreement with expectations based upon the bulk material parameters. In order to apply a similar analysis to the hexagonal phase, a finite number of approximations will be necessary. If we consider first the measurements on films deposited without subsequent annealing but showing clear presence for the hexagonal phase (Fig. 6) we observe a refractive index of 1.83 ± 0.02 . Assuming this film is comprised of amorphous and hexagonal Nd_2O_3 (there is no evidence in the X ray spectrum for the cubic phase) we deduce a film composition of 80% amorphous and 20% hexagonal. The measured dielectric constant was $12 - 12.7 \pm 0.2$ so that under the same

approximation of heterogeneity we conclude that the maximum dielectric constant of hexagonal Nd_2O_3 is ~ 21. Such a value is approximately consistent with the values of 20 – 27 reported for hexagonal La_2O_3 and 25 – 25.4 reported for hexagonal Pr_2O_3 (7).

If we analyse the measurements on annealed samples which appear to be almost "pure" hexagonal Nd_2O_3 (Fig. 6) the density was measured as 7.02 ± 0.2 g cm⁻³ whereas the bulk value is $7.14~{\rm g~cm}^{-3}$. Assuming the sample deposited at $280~{\rm °C}$ at a rate of 0.4

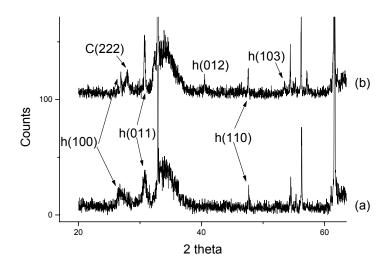


Fig. 6 X ray diffraction pattern for Nd₂O₃ deposited on Si(100) at 280 °C at 0.4 Å /sec. (a) followed by 600 °C annealing for 3h in O₂ and then at 800 °C for 30 min in N₂ (b). X ray lines are again identified with the different phases of the oxide: h for hexagonal. c for cubic. The position of the Si(200) which comes from the substrate is also indicated (lower curve). In the upper curve the sample was tilted with respect to the incidence direction of the X-rays to suppress the Si(200) reflection. The patterns show the dominance of the hexagonal phase in the as-deposited, 280 °C sample. After annealing both hexagonal and cubic phases, are present.

Å s⁻¹ then annealed in O_2 then N_2 is in fact the pure hexagonal phase, then the dielectric constant would be the measured value, $17\text{-}18 \pm 0.2$. Though larger than for the pure cubic phase, it is small when compared to hexagonal phases of La_2O_3 and Pr_2O_3 . Furthermore, the measured refractive index for this sample was $1.91 - 1.94 \pm 0.02$ which, compared to the hexagonal phase value of 2.11, suggests the film is not entirely pure hexagonal phase.

If we accept the principle of making an extremely naïve approximation we can use the data on cubic Nd_2O_3 to estimate $k_{hexagonal}$. From the Clausius-Mosotti equation:

$$(k-1)/(k+2) = 4 \pi \alpha_{dc}/3 V_m$$
 (2)

where α_{dc} is the low frequency molecular polarizability. Using the measured values of k_{cubic} (taken as 15) and the molecular volume of cubic Nd_2O_3 (85 ų) we determine $\alpha_{dc} \sim 16.7$ ų. Assuming this value appropriate for the hexagonal phase and taking $V_m(hexagonal) = 78$ ų we deduce $k_{hexagonal} \sim 27$. We reiterate that this simple calculation ignores volume dependence (if any) of α_{dc} and structural dependence i.e. we ignore any changes in α_{dc} which could result from differences in the vibrational spectrum of the two phases. Despite these approximations, $k_{hexagonal}$ is again suggested to be in the 20's.

Some support for this hypothesis may come from studying amorphous and hexagonal Pr_2O_3 where the low frequency polarizability is found to be network insensitive(5).

At the present time, then, there is a dilemma. Annealing of the deposited films at higher temperatures than 800 °C to ensure a maximized hexagonal phase content would lead to interfacial interaction with the substrate (3). There would appear to be no clear approach to forming the hexagonal phase on Si and profiting from its apparently higher dielectric constant than the cubic phase. One must thus conclude that such a material would therefore be impractical for technological purposes.

CONCLUSIONS

We have studied Nd₂O₃ films deposited under various conditions and subjected to different anneals. The dielectric constant of the cubic phase is determined to be in the range 13.6 - 15.4 which is consistent with measurements on other RE₂O₃ oxides. The refractive index at 632.8 nm wavelength is determined to be 1.93 ± 0.02 which is slightly less than the value of 1.97 we estimate on the basis of the Lorentz-Lorenz equation assuming $n_{\text{hexagonal}} \sim 2.11$ and taking account of differences in molecular volume. Hexagonal phase films were obtained by slow deposition at 280 °C and this result is surprising since this phase is not normally obtained until temperatures in excess of 850 °C are reached. This result strongly suggests that the substrate plays an important role in determining the structure of the deposited film. The highest refractive index we were able to obtain was 1.94 ± 0.02 which is significantly smaller than the anticipated hexagonal value and we have no explanation for this at the present time. We note that the film density was also smaller $(7.02 \pm 0.2 \text{ g cm}^{-3} \text{ as opposed to } 7.14 \text{ g cm}^{-3})$ but not significantly. Based on these numbers and measurements of the dielectric constant we determine that the value for the hexagonal phase lays in the range 17 - 21. Simple calculations based upon application of the Clausius-Mosotti equation using data for cubic Nd_2O_3 suggest $k_{hexagonal} \sim 27$. The major concern associated with the use of this material in technological applications would be a) the potential reactivity of Nd₂O₃ with the Si substrate and b) the apparent sensitivity of the structure of the deposited film to the deposition conditions (temperature and rate)..

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